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# Light cracked naphtha processing: Controlling chemistry for maximum propylene production

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## Abstract

As propylene market is expanding, new production paths have to be found. The cracking of light olefins contained in several naphthas seems to be a good alternative for responding to this demand. Results of light FCC naphtha cracking have shown that selectivity towards propylene is governed by hydrogen-transfer reactions. Thus, the selectivity to propylene may be increased by minimizing these reactions. This can be achieved by cracking the naphtha at high temperature, by using shape-selective catalysts or by working with coked catalysts. Recycling light naphtha in the FCC process is an interesting alternative, which may increase the yield of propylene by 50% if proper processing is carried out. Furthermore, olefins content of FCC gasoline may be significantly reduced.

*Keywords:* Naphtha; Cracking; Propylene; Olefin reduction

## 1. Introduction

Propylene is the key building block for the production of important petrochemicals, such as polypropylene, acrylonitrile, propylene oxide, cumene, phenol, isopropylic alcohol and many others [1]. As shown in Fig. 1, propylene worldwide demand has been increasing at an annual average of 5.7% since 1990; by year 2000, propylene production was about 52 million tonnes, and it is projected that by year 2010, demand will grow to 84 million tonnes, due to the increasing polypropylene demand [2].

Currently, about 70% of propylene worldwide production comes from steam crackers, 28% from refinery fluid catalytic cracking (FCC) units and 2% from specific on-purpose processes, such as propane dehydrogenation, metathesis and others. It is expected that in order to cover the future demand, the propylene market share from other processes than steam cracking will importantly grow, since it is forecast that propylene market will grow faster than ethylene market [2,3]. Although capacity of propylene on-purpose processes is going to be quadrupled in the next 5 years, it will still cover only a small part of the propylene demand, and other sources have to be developed. Thus, considering the worldwide FCC installed capacity (14 million barrels/day) and the already proven flexibility of the process to shift operation mode towards LPG olefins production, this process appears to be a sensible alternative that may contribute to satisfy the growing demand of this petrochemical.

It is well known that increasing process severity can significantly increase propylene production in FCC units. Likewise, propylene production may be increased by using olefins selective catalysts or by incorporating ZSM-5 zeolite-based additives into the catalytic system. Yet another emerging option to increase propylene yield is by reprocessing FCC naphtha. With this option, naphtha-range olefins may be transformed into LPG olefins, while at the same time, olefins content of FCC naphtha may be reduced, which would facilitate downstream upgrading [4–6].

The simplest option for naphtha processing in the FCCU is to add it directly to gas oil feed. Experimental results, however, showed that under this scheme and even with a catalyst system containing ZSM-5 zeolite, naphtha

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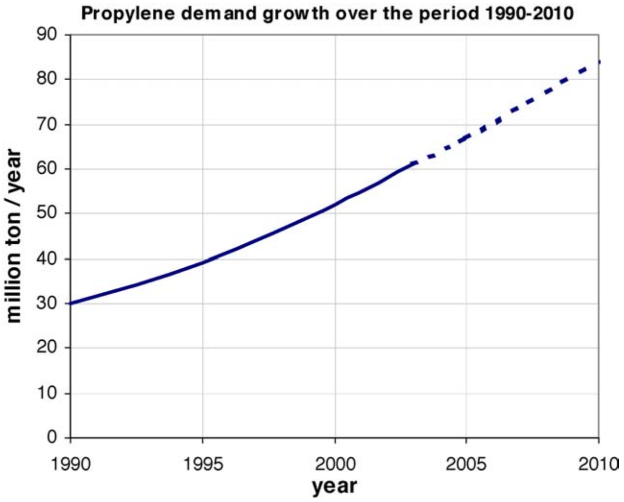

Fig. 1. Propylene demands expected to increase sharply in the next 10 years (1990–2010).

conversion was low [7] despite the high olefins content of the feedstock, and LCN only acted as a diluent of gas oil feed. This may be due to the higher adsorption ability of the gas oil molecules and polynuclear aromatic products. Thus, in order to effectively crack naphtha, it must be fed at a different location than gas oil in order to avoid competing effects. With the purpose to evaluate the potential of recycling FCC naphtha for increasing propylene yield, the following options (Fig. 2) were experimentally simulated in a transported bed MicroDowner (MD) reactor and a fixed bed (MAT) reactor:

(a) feeding naphtha at the bottom of the riser before the gas oil injection zone;
(b) processing naphtha in a parallel riser;
(c) processing naphtha in the stripper;
(d) processing in a separate process with a shape selective catalyst.

## 2. Experimental

The MD unit has been demonstrated to be able to satisfactorily reproduce operating conditions and yields of commercial units [8,9]. MAT unit follows features of ASTM 3907-86 but uses a modified test procedure and is fully automated [10,11]. Experiments were carried out over a mixture of 95 wt.% of a commercial low rare earth Y zeolite-based catalyst and 5 wt.% of ZSM-5 zeolite-based additive, using light FCC naphtha (LCN) and vacuum gas oil as feeds. Processing LCN was preferred over whole FCC naphtha, since the former fraction is richer in olefins, which are the most reactive and selective components to produce propylene, thus, making this alternative more cost-effective than processing the whole naphtha. Catalysts and feedstock properties are summarized in Table 1.

In some cases, a coked catalyst sample containing 1.2 wt.% of coke was used in MAT test. This was obtained by running a reaction with gas oil under standard reaction conditions. A time-on-stream (TOS) of 30 s, a catalyst-to-oil ratio (CTO) of 5 and a reaction temperature of 793 K. The amount of coke deposited in the catalyst is typical of an industrial operation.

Conversion is defined as the sum of dry gas, LPG and coke when cracking naphtha. In the case of simultaneous cracking of gas oil and naphtha, conversion is defined as the sum of liquid products boiling below 489 K, gases and coke.

Mass balances were considered acceptable when comprised in the limits 95–105% of injected feed.

## 3. Results and discussion

### 3.1. Reactions governing the cracking of naphtha

It has been well established that the conversion of light hydrocarbons occurs via carbenium ion chemistry. First, a carbenium ion is generated, either by addition of a proton on a double bond or by the addition of a proton on a saturated molecule, forming a pentacoordinated ion, which immediately cracks yielding a carbenium ion and a paraffin [12]. Then, the carbenium ion cracks yielding olefins and a smaller carbenium ion (monomolecular mechanism) or acts as an active site by reacting with a feed molecule (bimolecular mechanism) forming a larger molecule that further cracks. This second mechanism explains the formation of heavier compounds than those present in the

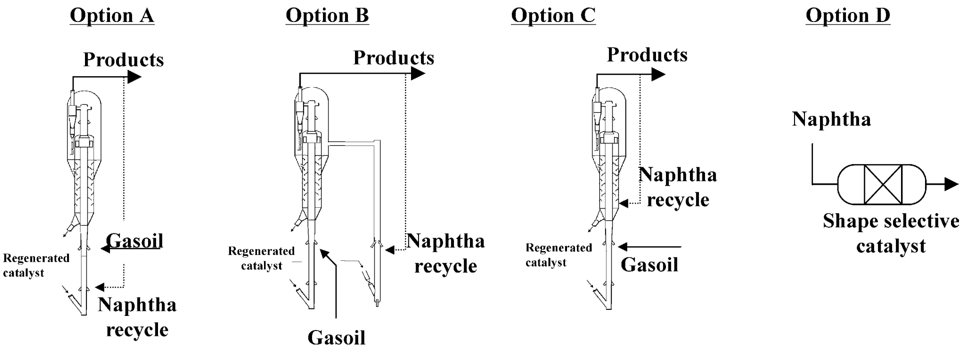

Fig. 2. Different schemes for naphtha recycling in the FCC.

Table 1
Catalysts and feedstock properties

| Catalysts | E-Cat | ZSM5-based additive | | | | | |
|---|---|---|---|---|---|---|---|
| Rare earth content (wt.%) | 1.0 | – | | | | | |
| Unit cell size (Å) | 24.30 | – | | | | | |
| Zeolite Si/Al ratio | 13 | 50 | | | | | |
| Specific area ($m^2/g$) | 114 | 70 | | | | | |
| Zeolite in catalyst (wt.%) | 11 | 15 | | | | | |
| Vacuum gas oil properties | | | | | | | |
| Specific gravity @ 60 °F | 0.917 | | | | | | |
| Average molecular weight (l b/mol) | 407 | | | | | | |
| $K_{UOP}$ | 11.82 | | | | | | |
| ASTM D1160 distillation | | | | | | | |
| In vol.% | 5 | 10 | 30 | 50 | 70 | 90 | 95 |
| In °F | 606 | 666 | 687 | 777 | 858 | 954 | 996 |
| Naphtha properties | LCN | | | | | | |
| Specific gravity @ 288 K | 0.68 | | | | | | |
| Average molecular weight | 84 | | | | | | |
| Composition, per hydrocarbon type (wt.%) | | | | | | | |
| Paraffins | 4.5 | | | | | | |
| Isoparaffins | 36.5 | | | | | | |
| Olefins | 43.8 | | | | | | |
| Naphthenes | 10.0 | | | | | | |
| Aromatics | 5.2 | | | | | | |

feed, as well as and the hydrogen-transfer reaction [13]. Finally, the carbenium ion can desorb as an olefin, thus, restoring the initial active site. This mechanism, called chain mechanism, has been useful to explain the conversion of small paraffins [14,15] and can be applied in the same way to the conversion of the other classes of hydrocarbons present in the naphtha feed, with the difference that, for olefins, the carbenium ion generation is much easier. The balance between mono- and bimolecular mechanism will depend on the surface coverage by carbenium ion. Thus, parameters, such as site density or temperature will have an influence on the cracking probability. Notice that site density may be decreased either by dealumination (irreversible) or coking (reversible) of the catalyst. On the other hand, a small pore size will favour the monomolecular mechanism since the bimolecular reaction intermediary cannot be formed in narrow pores, which was observed with the conversion of $C_5$–$C_9$ olefins on ZSM5 zeolite [16]. As an illustration, we can consider the conversion of naphtha range olefins. Following the chain mechanism paths, a single olefin can either generate a carbenium ion and crack into LPG-range olefins (monomolecular) or react with an existing carbenium ion (bimolecular). This second step can lead to a paraffin together with a more dehydrogenated carbenium ion (hydrogen transfer) or form a $C_{10}$–$C_{14}$ carbenium ion that can then cycle and lead after cracking and dehydrogenation to a $C_7$–$C_{11}$ aromatic and a corresponding amount of hydrogen or paraffins (dimerization-cracking).

The very high reactivity of olefins with respect to the corresponding paraffins under realistic FCC cracking conditions has been demonstrated [17]. Moreover, it was observed that the reactivity of olefins with six or less carbon atoms was considerably lower than the reactivity of longer chain olefins [18]. Respect to naphthenes, it has been shown [19] that methylcyclohexane preferentially crack towards smaller olefins, while methylcyclohexene, which can readily form the corresponding carbenium ion, preferably evolves towards aromatic products through hydrogen-transfer reactions or direct dehydrogenation. Thus, a bimolecular cracking mechanism will preferentially lead to the formation of aromatics from the naphthenes, since the carbenium may be directly formed from the feed molecule through a hydride transfer, while a monomolecular cracking mechanism will preferentially lead to short olefins. Then, the balance between hydrogen-transfer reactions and cracking reactions will be critical in orienting the selectivity of olefins and naphthenes conversion towards either propylene or gasoline-range aromatics and paraffins.

## 3.2. Light FCC naphtha cracking

### 3.2.1. Influence of temperature on the processing of light cracked naphtha

From Fig. 3, it is clear that at moderate temperature (723 and 823 K, transported bed), the hydrogen-transfer ability of the Y zeolite is the governing reaction, despite the presence of a significant amount of ZSM5 additive. This is reflected by the increase in the $C_5$ and $C_6$ paraffins. As a consequence, the yield of propylene is moderate (Table 2). The conversion of naphthenes is around 50–70%, and there is a parallel increase of the yield of aromatics. The higher carbon number distribution in the aromatics is well in accordance with the

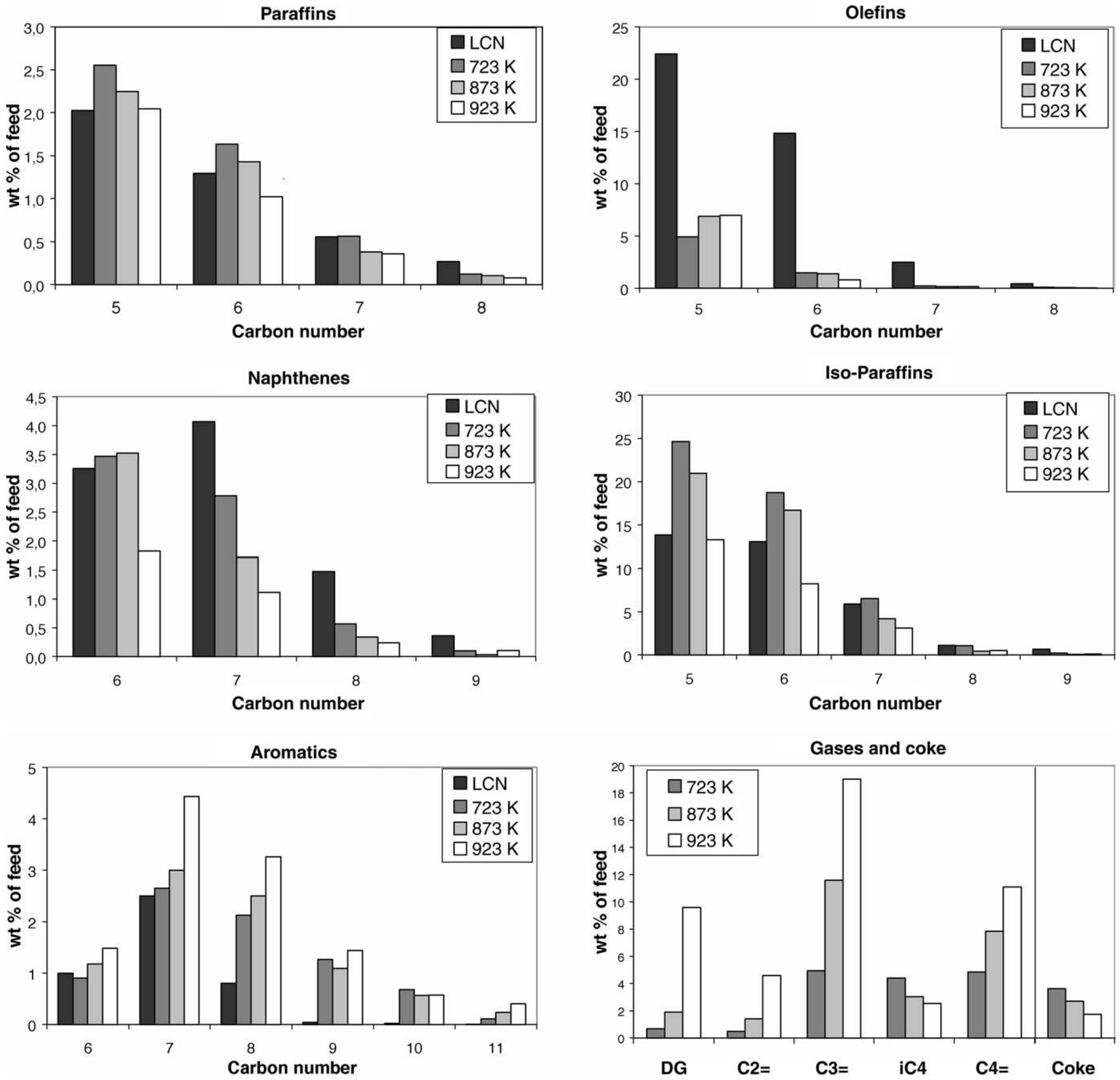

Fig. 3. Influence of the temperature on the selectivity of LCN cracking in the MicroDowner unit (transported bed), WHSV in the range 60–90 $h^{-1}$. From black to white: LCN feed, processing at 723, 823 and 923 K.

bimolecular reaction mechanism, where a feed molecule dimerizes with an adsorbed carbenium. At high temperature (923 K), the high propylene yield and the decrease of all paraffin yields indicate that cracking is the predominant reaction.

### 3.2.2. Influence of coke-on-catalyst

Comparing at the same space velocity, the presence of coke-on-catalyst reduces the cracking rate of $C_5$ olefins and gas and coke yields as presented in Table 3 and Fig. 4 (LCN 1 and 4). The same conversion towards gas and coke can be reached using a lower space velocity in a fixed bed (LCN 5), but the selectivity is different than those encountered with regenerated catalyst and transported bed. Paraffinic gasoline yield is lower and light olefins yield is higher with coke-on-catalyst, which clearly demonstrates that it reduces the ratio of hydrogen-transfer reaction rate over cracking reaction rate. This can be achieved in two ways. The Y zeolite may be more deactivated than the ZSM5 zeolite, thus, favoring the conversion of the LCN on this late, more selective to cracking [20]. Also, the coke may decrease the site density on Y zeolite, which will decrease the reaction rate for bimolecular reactions like hydrogen transfer [21,22]. It is worth to notice that the Y zeolite still have enough activity

Table 2
Operating conditions and product yields for LCN cracking in the MicroDowner unit (transported bed) at various temperatures

| Test name | LCN 1 | LCN 2 | LCN 3 |
|---|---|---|---|
| Temperature (K) | 723 | 823 | 923 |
| Space velocity ($\mathrm{h^{-1}}$) | 68 | 72 | 91 |
| Conversion (wt.%) | 19.3 | 27.8 | 46.1 |
| $C_2^=$ | 0.49 | 1.42 | 4.60 |
| Dry gas | 0.69 | 1.92 | 9.58 |
| $C_3$ | 0.36 | 0.65 | 1.47 |
| $C_3^=$ | 4.93 | 11.58 | 19.02 |
| $iC_4$ | 4.41 | 3.04 | 2.55 |
| $nC_4$ | 0.45 | 0.43 | 0.61 |
| $iC_4^=$ | 1.66 | 2.94 | 4.02 |
| Total $C_4^=$ | 4.83 | 7.85 | 11.09 |
| $C_3^=/\text{total}\,C_3$ | 0.93 | 0.95 | 0.93 |
| $iC_4^=/iC_4$ | 0.38 | 0.97 | 1.58 |
| Coke | 3.62 | 2.30 | 1.74 |
| Coke-on-catalyst | 0.11 | 0.07 | 0.05 |

for transforming a significant part of feed olefins into $C_5$–$C_6$ paraffins through hydrogen-transfer reaction despite a coke-on-catalyst content of 1.2 wt.%

### 3.2.3. Conversion of LCN on shape-selective ZSM5-based additive

In this case, the operating conditions may be varied in a wide range, as they do not depend on FCC process constraints due to simultaneous cracking of gas oil. The LCN was processed at 823 K in order to maximize the selectivity to light olefins but with a limited production of dry gases through thermal cracking reactions, especially methane and ethane. As presented in Table 4, a high yield of propylene, nearly, 20% by weight can be achieved. The dry gas yield is high due to the catalytic production of ethylene, but is still lower than this obtained at 923 K with a similar yield of propylene (LCN 3 in Table 2). The very large difference in the propylene yield with the commercial mix of catalyst (test LCN 2) shows that nearly half of the potential light olefin production is lost through hydrogen-transfer reactions of feed olefins and naphthenes. Despite the high content of olefins in the feedstock, coke yield is extremely low. This allows considering a fixed bed process for the conversion of the naphtha with periodical regeneration. The rapid conversion of olefins allows a limited size of the catalytic bed for the operation. As showed in Table 4, a lower space velocity than 100 $\mathrm{h^{-1}}$ do not improve substantially the yields of LCN processing. In fact, it has already been demonstrated that the conversion of LSR naphtha ($C_5$–$C_7$ paraffins) at this temperature is not of interest.

Table 3
Influence of coke yield and catalyst contact time on cracking selectivity of LCN in MicroDowner (transported bed) and MAT unit (fixed bed)

| Test name | LCN 1 | LCN 4 | LCN 5 |
|---|---|---|---|
| Type of bed | Transported | Fixed | Fixed |
| Catalyst | Regenerated | Coked | Coked |
| Temperature (K) | 723 | 723 | 723 |
| Space velocity ($\mathrm{h^{-1}}$) | 68 | 60 | 12 |
| Conversion (wt.%) | 19.3 | 12.1 | 20.2 |
| $C_2^=$ | 0.49 | 0.21 | 0.45 |
| Dry gas | 0.69 | 0.29 | 0.60 |
| $C_3$ | 0.36 | 0.13 | 0.31 |
| $C_3^=$ | 4.93 | 4.37 | 6.9 |
| $iC_4$ | 4.41 | 1.08 | 2.34 |
| $nC_4$ | 0.45 | 0.18 | 0.30 |
| $iC_4^=$ | 1.66 | 3.12 | 3.85 |
| Total $C_4^=$ | 4.83 | 6.08 | 8.56 |
| $C_3^=/\text{total}\,C_3$ | 0.93 | 0.96 | 0.96 |
| $iC_4^=/iC_4$ | 0.38 | 2.97 | 1.64 |
| Coke | 3.62 | 0.75 | 1.22 |
| Coke-on-catalyst | 0.11 | 0.15 | 0.20 |

## 3.3. Estimation of gas oil cracking yields with naphtha recycle

The yields presented in Table 5 are the result of combining gas oil cracking (in the first column, base case) and LCN processing yields following the schemes presented in Fig. 2. This calculation was carried out by subtracting an amount equivalent to recycled naphtha (21% of gas oil feed) from base case gasoline yield and adding the naphtha cracking yields (referred to 21% of the gas oil feed) to the base case yields. The LCN cracking tests used for each of the processing scheme were: LCN 2 and 3 for the simulation of parallel riser and the cracking at the bottom of the riser, respectively, LCN 5 for the simulation of cracking in the stripper and LCN 7 for the simulation of a separate process. The base case already considers a high LPG olefins yield through ZSM5 addition.

### 3.3.1. Cracking of naphtha before gas oil injection

A practical option for processing LCN separately from gas oil feed would be to inject it into the bottom of the riser, since this implies few hardware modifications (i.e. adding a naphtha feeding system). In this case and if all LCN were recycled, cracking would be carried out at very high temperature (>920 K) and catalyst-to-feed ratio (CTO > 30), since the amount of naphtha recycled represents only a small fraction of the gas oil feed (20–25 wt.% maximum). The low coke yield generated in the LCN cracking, only about 0.06 wt.% coke-on-catalyst, will not affect the further gas oil conversion. As shown in Table 2, in the case that all LCN was recycled, propylene production increased by almost 50% over the base case, which for a 40,000 barrels/day (5760 tonnes/day) FCC unit would mean an additional propylene production equivalent to 240 tonnes/day, besides, about 130 tonnes/day of butylenes would be also produced. The selectivity towards LPG olefins referred to naphtha loss was around 70%. An additional benefit would be the reduction of more than 50% olefins in the resulting FCC naphtha, whose content would be of only 12 wt.% as can be seen in Table 3.

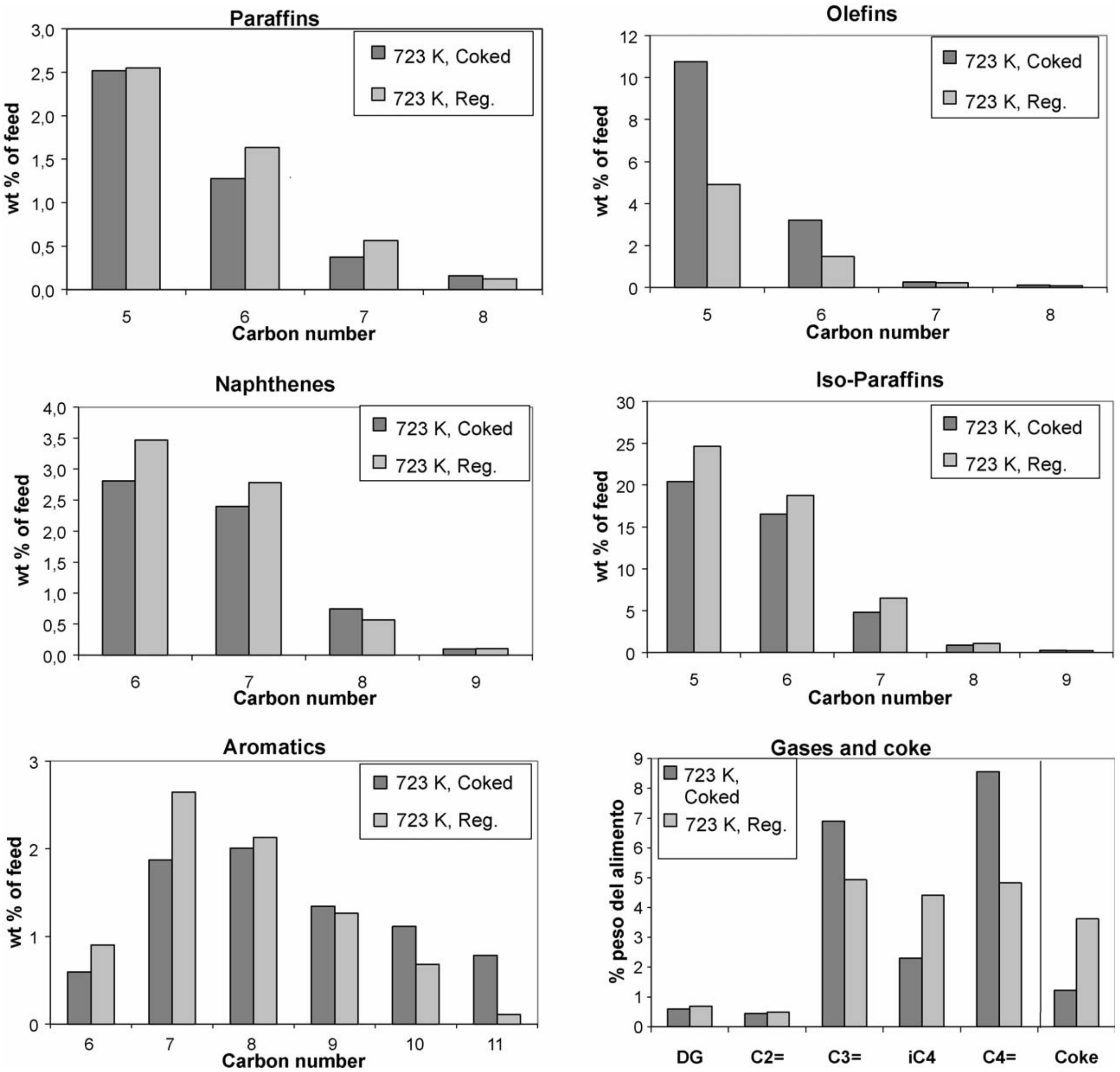

Fig. 4. Influence of coke-on-catalyst on the activity and selectivity of LCN cracking at 723 K, at a conversion around 20% (tests LCN 1 and 5, Table 3).

The major operational drawback of this operation would be the substantial increase in dry gas yield resulting from thermal cracking, which was about 80% higher than the base case, making necessary to increase the gas compressor capacity.

### *3.3.2. Cracking of naphtha in an independent riser*

Due to the high dry gas yield reported by the previous scheme, it would be desirable to convert LCN at lower temperature to minimize thermal reactions. This could be achieved by processing LCN in a separated riser, parallel to gas oil cracking riser. Indeed, simulation of this case (LCN 2) showed that dry gas yield increment was only 32% higher than the base case and sensitively lower than in the previous case.

Although LCN conversion to gas and coke resulted smaller because of the lower reaction severity, gasoline-range olefins conversion is only a little bit smaller than in the previous option, resulting in the same interesting reduction of olefins in the gasoline. As the selectivity in the conversion of the olefins was quite different, the yield of propylene increased only by 25% over the base case as shown in Table 5, while the selectivity towards LPG olefins was similar to the previous case. As an advantage, this scheme allows a more flexible control of reaction temperature and cat/oil to achieve the propylene production desired within the range allowed by LCN conversion. The major drawback is the higher investment, operation and maintenance cost derived from the installation of a new riser attached to the gas oil riser.

Table 4
Cracking yields with pure ZSM5 additive at 823 K obtained with the MicroDowner unit

| Test name | LCN 6 | LCN 7 |
|---|---|---|
| Temperature (K) | 823 | 823 |
| Space velocity ($h^{-1}$) | 105 | 58 |
| Conversion (wt.%) | 39.1 | 41.7 |
| $C_2^=$ | 5.62 | 6.54 |
| Dry gas | 5.96 | 6.93 |
| $C_3$ | 0.91 | 1.15 |
| $C_3^=$ | 18.53 | 19.53 |
| $iC_4$ | 0.48 | 0.59 |
| $nC_4$ | 0.38 | 0.46 |
| $iC_4^=$ | 4.96 | 4.97 |
| Total $C_4^=$ | 12.45 | 12.51 |
| $C_3^=/\text{total } C_3$ | 0.95 | 0.94 |
| $iC_4^=/iC_4$ | 10.41 | 8.41 |
| Coke | 0.34 | 0.47 |
| Coke-on-catalyst | 0.01 | 0.01 |

### *3.3.3. Cracking of naphtha in the stripper*

Although this is not the best option for producing propylene, processing LCN in the catalyst stripper is an attractive alternative if gas compressor is a constraint, as dry gas increase over the base case is very low. LCN conversion is lower than in the other options while the selectivity towards light olefins is slightly higher. Furthermore, the olefin content of resulting FCC naphtha is reduced to about one half of the base case while aromatic and *iso*-paraffinic content of cracked naphtha present the lowest changes. Thus, this would be the preferred option if the main goal were to upgrade FCC naphtha quality with an appreciable propylene and butylenes production as a collateral benefit.

### *3.3.4. Cracking of naphtha in a separate process*

This option offers the highest selectivity to propylene and butenes with a slightly lower dry gas generation than the best former option for propylene production (bottom of the riser cracking option). Moreover, the dry gas increase is due to ethylene only, which can also be considered as a valuable product. The LPG olefins yield increases a 35% over the base case. Nevertheless, this option is subjected to a carefully economic evaluation because of the huge inversion and the fact that most of the heat required by the process should be generated independently, since the coke make is very low and an autothermal operation like FCC operation would not be possible.

It is worth to notice that in all the processing options considered, the aromatic content of total gasoline is significantly increased, because of the concentration of the heavy gasoline-range aromatics in the total gasoline, resulting in an octane increase of the gasoline. The highest octane gain will be with the most severe process, as the concentration effect is the highest due to the highest LCN conversion.

## 4. Conclusion

It has been shown that propylene yield from FCC units can be increased far beyond current values by reprocessing the light FCC naphtha cut, leading to a yield of 25% weight of LPG olefins from gas oil processed with current FCC technology. This may be achieved through different schemes, among which those that maximize propylene yield require some major modification; naphtha cracking at the bottom of the riser demands for higher compressor capacity and naphtha processing in a separate process

Table 5
FCC products yields comparison

| LCN processing catalyst | Y + 5 wt.% ZSM5 | | | | ZSM5 |
|---|---|---|---|---|---|
| | None | Bottom of the riser | Parallel riser | Separator | Separate process |
| Products yield (wt.%) | | | | | |
| Dry gas | 2.4 | 4.4 | 2.8 | 2.5 | 3.8 |
| LPG | 23.6 | 30.8 | 28.2 | 27.3 | 30.7 |
| Propene | 8.6 | 12.5 | 10.6 | 10.0 | 12.7 |
| Butenes | 10.6 | 12.9 | 12.2 | 12.5 | 13.2 |
| Gasoline | 47.3 | 37.7 | 41.9 | 43.1 | 38.6 |
| Coke | 3.9 | 4.2 | 4.4 | 4.3 | 4.0 |
| Selectivity of LCN cracking to | | | | | |
| Olefins LPG | | 65 | 67 | 79 | 77 |
| Propene | | 40 | 37 | 33 | 47 |
| Composition of the gasoline | | | | | |
| Paraffins | 33 | 35 | 41 | 40 | 39 |
| Olefins | 25 | 12 | 11 | 14 | 11 |
| Naphthenes | 10 | 9 | 10 | 10 | 10 |
| Aromatics | 32 | 44 | 39 | 37 | 40 |

Projected results @ 77% gas oil conversion, with a recycle of LCN equivalent to 21 wt.% of the gas oil feed.

requires a huge inversion and an independent heat supply. In contrast, LCN processing in the stripper also significantly increases propylene production without requiring major modifications on the unit, having the advantage of producing only small quantities of additional dry gas and significantly lowering olefin content of cracked naphtha.

It is worth to mention that in all cases LCN naphtha is converted with higher selectivity to LPG olefins than could do a steam cracking operation, being the average value around 70 wt.%, and it is 35 wt.% for propylene.

The different proposed cracking schemes offer the refiner a range of options for increasing propylene production depending on capital available and spot market economics. Further increase of propylene yield over the results presented could be achieved by increasing severity of the gas oil conversion process.

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